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Publisher's version / Version de l'éditeur:

<https://doi.org/10.1016/j.combustflame.2013.02.007>

Combustion and Flame, 160, 9, pp. 1693-1705, 2013-06

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Soot Temperature and Volume Fraction Retrieval from Spectrally Resolved Flame Emission Measurement in Laminar Axisymmetric CoFlow Diffusion Flames: Effect of Self-Absorption

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Abstract

The effects of self-absorption on the inverted soot temperature and volume fraction distributions in coflow laminar diffusion flames were investigated using two correction methods. Self-absorption was found unimportant when the optical thickness of the flame is less than 0.3, but can be very important in flames of large optical thicknesses. Due to the uncertainty in soot absorption function $E(m)$, different assumptions of the variation of $E(m)$ with wavelength were investigated. Decreasing $E(m)$ with wavelength leads to largest errors in the recovered soot temperature and volume fraction when self-absorption correction is not accounted for. The assumption of constant $E(m)$ in general results in smallest errors in the inverted soot volume fraction and the increasing $E(m)$ with wavelength gives rise to smallest errors in the recovered soot temperature. Neglect of self-absorption correction in general leads to underestimate of the inverted soot temperature, while the inverted soot volume fraction can be higher or lower than the true value, depending on how $E(m)$ varies with wavelength. The self-absorption correction method of Snelling et al. [AIAA J. 2002] is effective in recovering accurate soot temperature and volume fraction distributions from noise-free emission data even for flames with optical thickness as large as 5. For inversion of emission data containing noise it is still advantageous to account for self-absorption. Multi-wavelength emission data from a laminar coflow ethylene/air diffusion flame at atmospheric pressure and a methane/air diffusion flame at 40 atm were reanalyzed to demonstrate how self-absorption affects the reconstructed temperature and soot volume fraction distributions. It is preferred to detect flame emissions at longer wavelengths in the visible spectrum to minimize the self-absorption effect. The effect of self-absorption should in general be taken into account in the reconstruction of soot temperature and volume fraction from the line-of-sight spectral flame emission data using the method of Snelling et al.

Key Words: Soot measurement; line-of-sight spectral flame emission; self-absorption; Abel inversion

1. Introduction

With the advance of our knowledge on the adverse impact of soot on environment, climate, and human health, increasingly stringent regulations have been enforced in many regions and countries to limit the amount of soot emissions from various combustion devices, such as automobiles and aircrafts. This situation has generated renewed interests in soot study to gain fundamental understanding of soot formation and oxidation mechanisms and help search for effective strategies to mitigate soot emissions from practical combustion devices. Optical diagnostic techniques play an important role in improving our understanding of soot formation and in the evaluation of combustion technologies in terms of soot formation and emissions. Various optical diagnostic methods for soot temperature, volume fraction, and morphology measurements have been developed in the last several decades, such as light extinction for concentration [1,2], flame emission for soot temperature [3,4], the coupled light extinction and flame emission [5] for soot concentration and temperature, multi-wavelength flame emission detections [6-8] for soot concentration and temperature, laser elastic scattering [9,10] for soot aggregate size distribution, and laser-induced incandescence [11,12] for soot volume fraction and primary particle size. The steady-state laminar axisymmetric coflow diffusion flame is an important flame configuration to study soot formation mechanisms since it is stable and highly reproducible and allows for laser based non-intrusive diagnostics for detailed characterizations of soot.

For investigation of soot volume fraction distributions in laminar axisymmetric coflow diffusion flames both emission-absorption tomography and multi-wavelength soot emission in the visible spectrum tomography have been used, e.g. [5-8, 13-16], to obtain radially resolved soot temperature and volume fraction distributions. These techniques are based on the fact that soot is a strong absorber and emitter of light in the visible spectrum with fairly known optical properties and offer the advantages of rapid time response, being non-intrusive, economic, and easy to set up. As in almost all optical diagnostics (except light extinction), flame emission measurements also suffer the effect of signal trapping or self-absorption, i.e., the thermal emission intensity from a small volume along the path between this volume and the detector is attenuated (absorbed and scattered) by soot present between this volume and the detector. Self-absorption always reduces the signal strength reaching the detector and can be significant under conditions of large optical thicknesses between the signal source and the detector. Considerable research attention has been paid to self-absorption, or signal trapping, in soot volume fraction measurement using LII in axisymmetric laminar sooting flames, e.g. [11,17-21]. The more recent studies on correction of LII signals due to self-absorption [18-21] followed the methodology proposed by Choi and Jensen [17]. Almost all these studies employed the Beer-Lambert exponential extinction law to recover the actual, unattenuated LII signals, with the exception of the studies of Liu et al. [21] and Daun et al. [22] who took into account the multi-dimensional effect of soot scattering on the LII signals reaching the detector by solving the radiative transfer equation using the discrete-ordinates method and a backward Monte Carlo method, respectively. Although scattering was found unimportant to signal extinction in typical laminar diffusion flames at atmospheric pressure in these studies, these methods can be readily applied to other situations where the effect of scattering can be important.

Compared to the studies of the self-absorption effect in LII measurements, there have been fewer efforts to correct the self-absorption effect in multi-wavelength flame emission tomography for soot measurements. A serious problem caused by the self-absorption phenomenon in multi-wavelength flame emission tomography in axisymmetric flames is that the resultant line-of-sight emission intensity expression is no longer an Abel integral equation and poses additional difficulties in data inversion to obtain radially resolved soot temperature and volume fraction. Consequently, various Abel inversion methods developed for the Abel integral equation cannot be readily used to invert the detected line-of-sight flame emission intensities for radially resolved quantities, unless the self-absorption effect is neglected. The strategy for self-absorption correction is to recover the unattenuated line-of-sight emission intensity, which is expressed as an Abel integral equation. Freeman and Katz [23] proposed a simple correction method to recover the unattenuated line-of-sight emission intensities based on a two-path approach. Their method was applied by Best et al. [24] to account for the self-absorption effect in the measured flame radiance data for reconstruction of radially resolved soot temperature in a laminar axisymmetric coflow ethylene/air diffusion flame. The potential importance of self-absorption of soot emission in axisymmetric laminar diffusion flames was also recognized by Hall and Bonczyk [5] in emission-absorption tomography and by Snelling et al. [7] in spectrally resolved flame emission measurement for soot temperature and volume fraction reconstruction. Both Hall and Bonczyk [5] and Snelling et al. [7] proposed a method to account for the effect of self-absorption by correcting the detected flame radiation intensity to recover the unattenuated value. Neither the method of Hall and Bonczyk nor the method of Snelling et al. has been validated or systematically used to investigate the self-absorption effect in the retrieval of soot temperature distribution in laminar axisymmetric coflow diffusion flames. Snelling et al. [7]

applied their correction method to account for the effect of self-absorption on the inverted radial distributions of soot temperature and volume fraction at a height of 3 cm above the burner exit surface of an atmospheric pressure laminar coflow ethylene/air diffusion flame. Their results showed that the inverted soot temperatures become only slightly higher (about 10 K) and soot volume fraction remain almost unchanged when self-absorption was taken into account, i.e., the influence of self-absorption in this flame was found very small. More recently Sun et al. [25] treated the data inversion of multi-wavelength flame emission measurement as an optimization problem and applied the stochastic particle swarm optimization (SPSO) method to simultaneously recover soot temperature and volume fraction distributions in the coflow laminar ethylene/air diffusion flame studied by Snelling et al. [7]. The experimental radial profiles of soot temperature and volume fraction of Snelling et al. [7] at two flame heights of 30 and 50 mm above the burner exit were used as input to simulate the line-of-sight spectral emission intensities over the spectral range of 500 to 1000 nm. In the study of Sun et al. [25], a constant soot refractive index of $m = 1.57 + 0.56i$ was assumed in their analysis, which corresponds to a soot absorption function $E(m) = 0.26$. SPSO was shown to be able to recover the input soot temperature and volume fraction distributions at the two flame heights of 30 mm and 50 mm with relatively good accuracy when the signal-to-noise ratio of the simulated emission data is very high. To highlight the effect of self-absorption on the reconstructed soot temperature and volume fraction distributions at large optical thickness, they multiplied the measured soot volume fractions of Snelling et al. [7] by a factor of 10 and showed that neglect of self-absorption leads to underestimate of soot temperatures by about 200 K. However, the effect of assuming different spectral variations of $E(m)$ was not investigated and how self-absorption affects the inverted soot temperature and volume fraction distributions were not explained.

Moreover, SPSO suffers the drawback of being computationally expensive to achieve convergence.

There is currently a lack of clear understanding on how self-absorption affects the reconstructed soot temperature and volume fraction distributions under different soot volume fraction distributions and different assumptions of the relationship between soot absorption function $E(m)$ and wavelength. The present numerical study was motivated by this observation and made an attempt to gain such understanding. In this study the two-path method of Freeman and Katz [23] and the emission intensity correction method of Snelling et al. [7] were employed to simultaneously reconstruct the radial distributions of soot temperature and volume fraction based on multi-wavelength flame emission measurement. The methods were first validated by comparing the recovered radial distributions of soot temperature and volume fraction with known ones reported by De Luliis et al. [6] in a laminar ethylene diffusion flame. Three different assumptions of the variation of $E(m)$ with wavelength were considered: decreasing, constant, and increasing. The effectiveness of the two methods and the importance of self-absorption correction in flames of large optical thickness were also demonstrated by increasing the input soot volume fraction distributions in the laminar ethylene diffusion flame studied by De Luliis et al. [6] by a factor up to 10. The multi-wavelength emission data from the laminar coflow ethylene/air diffusion flame at atmospheric pressure [6] were reanalyzed to investigate how the self-absorption effect affects the reconstructed soot temperature and volume fraction distributions. The spectrally resolved flame emission data of Thomson et al. [13] obtained from a laminar coflow methane/air diffusion flame at 40 atm were also reanalyzed with and without self-absorption correction to further demonstrate its effect. The roles played by the range of

detection wavelength in the effects of self-absorption on the reconstructed soot temperature and volume fraction distributions were also discussed.

2. Theory

We consider spectral radiation emission from soot over a range of wavelength in the visible spectrum in an axisymmetric laminar flame transmitted along a line-of-sight to reach a spectrometer, as schematically shown in Fig. 1. A scan of the spectrometer across the width of the flame (direction y) provides a set of projections consisting of line-of-sight radiation intensities at different y locations from $-R$ to R (R is the visible flame radius defined as the radial location where soot vanishes at the height under consideration) and at different wavelengths. Further details on typical optics and detectors used in the method can be found in [7]. Once the detection system is properly calibrated, the detected spectral line-of-sight radiation intensities can be related to the unknown soot temperature and volume fraction distributions provided that the optical property of soot is assumed known.

In this study, as in almost all previous studies dealing with emission and transmission of light in laminar diffusion flames, it is assumed that the flame medium is only absorbing and emitting, but non-scattering. This assumption is supported by the findings of a recent study of the relative importance of absorption and scattering to signal trapping in LII measurement in a laminar coflow ethylene/air diffusion flame [21]. Under this assumption, the line-of-sight radiation intensity detected by the spectrometer at a wavelength λ and a distance y from the flame center can be written as

$$I_{\lambda}(y) = \int_{-\sqrt{R^2-y^2}}^{\sqrt{R^2-y^2}} K_{a,\lambda}(x,y) B_{\lambda}[T(x,y)] \exp\left(-\int_x^{\sqrt{R^2-y^2}} K_{a,\lambda}(x',y) dx'\right) dx \quad (1)$$

where x represents the spatial location along the line-of-sight under consideration, $K_{a,\lambda}$ is the soot absorption coefficient and is related to the soot volume fraction f_v , wavelength λ , and the soot absorption function $E(m_\lambda)$ using the Rayleigh theory as

$$K_{a,\lambda}(x,y) = \frac{6\pi E(m_\lambda) f_v(x,y)}{\lambda} \quad (2)$$

The soot absorption function $E(m_\lambda)$ is related to the soot refractive index m_λ as $E(m_\lambda) = -\text{Im}[(m_\lambda^2 - 1)/(m_\lambda^2 + 2)]$. The blackbody spectral radiation intensity is expressed as $B_\lambda[T(x,y)] = 2\pi hc^2 \lambda^{-5} [\exp(hc/k\lambda T) - 1]^{-1}$. In this expression, h , c , and k are the Planck constant, speed of light, and the Boltzmann constant, respectively. In Eq. (1), the term $K_{a,\lambda}(x,y) B_\lambda[T(x,y)]$ represents the local spectral emission rate $S_\lambda(r)$ and the exponential term represents the transmission of the local emission through the region between the local emission volume and the spectrometer, i.e., this term reflects the self-absorption effect. Because of the presence of the exponential term, Eq. (1) is no longer an Abel integral equation and the inversion methods developed for Abel integral equation cannot be applied to invert Eq. (1) to obtain the local spectral emission rate distribution. Two methods to recover the local spectral emission rates will be described below. Assuming the local spectral emission rates $S_\lambda(r)$ can be determined, the following relationship can be obtained from the definition of $S_\lambda(r)$ and the expressions for the soot absorption coefficient and the blackbody spectral radiation intensity, following [7]

$$\ln \frac{E(m_\lambda)}{S_\lambda \lambda^6} = \frac{hc}{kT} \frac{1}{\lambda} - \ln(12\pi^2 hc^2 f_v) \quad (3)$$

In deriving Eq. (3) the Wien approximation of the blackbody function was invoked, which is very accurate in the visible spectral range under the flame temperatures. The local soot temperature can be recovered from the slope of $\ln[E(m_\lambda) \times S_\lambda(r)^{-1} \times \lambda^{-6}]$ vs. λ^{-1} at a given radial

location. Once the local emission rate $S_\lambda(r)$ and soot temperature are available, the local soot absorption coefficient is known. The local soot volume fraction can then be obtained from the local soot absorption coefficient given in Eq. (2) as

$$f_v = \frac{\lambda S_\lambda}{6\pi E(m_\lambda) B_\lambda(T)} \quad (4)$$

It is noted that at a given radial location the soot volume fraction should be independent of wavelength. Yet, the right hand side of Eq. (4) is in general slightly dependent on wavelength in numerical calculations, especially when self-absorption correction is not accounted for. To eliminate the wavelength dependence of soot volume fraction calculated using Eq. (4), the average value of soot volume fractions calculated from Eq. (4) over all the wavelengths at a given radial location is used to represent the local soot volume fraction.

The multi-wavelength flame emission technique for soot T and f_v measurement requires the soot refractive index m_λ over the spectral range of the detection. This quantity is subject to somewhat large uncertainty and is the topic of ongoing debate and research. As our focus is the effect of self-absorption, it is assumed that $E(m_\lambda)$ is known. However, the effect of making different assumptions of the $E(m_\lambda)$ vs. wavelength relationship will be investigated. Although the optical property of soot may vary with location in a flame as a result of different growth history and different H/C ratio, such spatial variation of $E(m_\lambda)$ is neglected in this study, which is a common practice in optical based soot measurements.

Two methods to account for the effect of self-absorption in the detected spectral radiation intensities are discussed below. The strategy of both methods is to correct the detected (attenuated) radiation intensity in order to recover the unattenuated value. The unattenuated line-of-sight spectral radiation intensity can be written as

$$I_{\lambda}^u(y) = \int_{\sqrt{R^2-y^2}}^{\sqrt{R^2-y^2}} K_{a,\lambda}(x,y) B_{\lambda}[T(x,y)] dx \quad (5)$$

Eq. (5) is a standard Abel integral equation and can be readily inverted by any inversion technique to obtain the local emission rate $S_{\lambda}(r) = K_{a,\lambda}(\tau) B_{\lambda}[T(r)]$.

2.1 The two-path method of Freeman and Katz

The starting point of the two-path method of Freeman and Katz [23] is to formulate the radiation intensity contributions of a differential emission volume on a given line-of-sight to two detectors placed in opposite directions. By assuming the optical thickness is small, the exponential term can be linearized. Integration of the differential contribution to the two detectors over the entire line-of-sight should be identical since the flame is axisymmetric. These procedures lead to the following relationship between the attenuated and unattenuated line-of-sight radiation intensities [23]

$$I_{\lambda}^u(y) = \frac{2}{1 + \tau_{\lambda}(y)} I_{\lambda}(y) \quad (6)$$

where $\tau_{\lambda}(y)$ is the spectral transmissivity along the line-of-sight at a distance of y from the flame center, i.e., $\tau_{\lambda}(y) = \exp[-\int_{\sqrt{R^2-y^2}}^{\sqrt{R^2-y^2}} K_{a,\lambda}(x,y) dx]$.

2.2 The correction method of Snelling et al.

To obtain the unattenuated line-of-sight spectral radiation intensity Snelling et al. [7] proposed the following simple method to correct the attenuated detected intensity

$$I_{\lambda}^u(y) = \frac{I_{\lambda,calc}^u(y)}{I_{\lambda,calc}(y)} I_{\lambda}(y) \quad (7)$$

where $I_{\lambda, \text{calc}}^u(\nu)$ and $I_{\lambda, \text{calc}}(\nu)$ are the calculated line-of-sight spectral radiation intensities without and with self-absorption using Eqs. (5) and (1), respectively. The method is based on the fact that the ratio of the unattenuated to attenuated line-of-sight spectral emission intensity can be simulated for known distributions of soot temperature and volume fraction. It is noted that in Eq. (7) $I_{\lambda}(\nu)$ is the only measured quantity (attenuated line-of-sight spectral emission intensity).

Since T and f_v distributions are unknown *a priori* but are required to calculate $\tau_{\lambda}(\nu)$ in Eq. (6) and $I_{\lambda, \text{calc}}^u(\nu)$ and $I_{\lambda, \text{calc}}(\nu)$ in Eq. (7), both the Freeman and Katz method and the correction method of Snelling et al. recover the soot temperature and volume fraction distributions through iterations. A good initial estimate of soot temperature and volume fraction distributions is that from the inversion of the measured data with self-absorption effect neglected, i.e., by setting the exponential term in Eq. (1) to unity. Because the Freeman and Katz method is based on linearization of the exponential term, it is expected that this method is accurate for small optical thicknesses. The method of Snelling et al., however, is not restricted to problems of small optical thickness.

3. Results and Discussion

Numerical calculations of the line-of-sight spectral radiation intensities and the inversion of the Abel integral equation, Eq. (5), were conducted by dividing the flame cross-section into a number of equally-spaced rings as shown in Fig. 1. All the results presented below were obtained by the three-point Abel inversion algorithm of Dasch [26] to obtain the local spectral emission rate. Calculations of the Snelling et al. [7] method and the Freeman and Katz method [23] started with the estimated T and f_v obtained by setting the exponential term to unity in Eq. (1). Unless

otherwise indicated, iteration was stopped when the maximum relative change in temperature between two successive iterations is less than 2×10^{-4} .

3.1 *Effects of self-absorption for decreasing $E(m_\lambda)$ with wavelength*

The two methods for accounting for the effect of self-absorption were first used to recover known distributions of soot temperature and volume fraction and to investigate the effect of self-absorption at different flame heights where soot concentration displays different distributions. Comparison of the recovered distributions to the known ones also serves as validation of the two methods. This is a commonly used method to check the accuracy of an inverse method, e.g., [27]. For these purposes, we used the T and f_v distributions in a laminar coflow ethylene/air diffusion flame at atmospheric pressure reported by De Iuliis et al. [6]. The T and f_v distributions at three flame heights of $z = 3, 4$, and 5 cm above the burner exit given in Figs. 5 and 3 of [6] were used as known soot temperature and volume fraction distributions. It is recognized that the soot volume fraction distributions given in Fig. 3 of [6] were obtained from extinction measurements and those from multiwavelength emission measurements were given in Fig. 4 of [6]. The reasons for using the f_v distributions given in Fig. 3 of [6] in our present study to investigate the effect of self-absorption in flame emission measurements are based on the following two considerations. First, the extinction measurements in [6] had a higher spatial resolution than the emission measurements. Second, there is good agreement between the results of extinction measurement and emission measurement after reducing the spatial resolution of the extinction results [6]. In [6], the line-of-sight spectral radiation intensity was detected between 300 and 800 nm at a spectral resolution of 25 nm, giving a total of 20 spectral intervals. In all the results reported in this section, the soot refractive index takes a wavelength dependent form as

suggested by Chang and Charalampopoulos [28], yielding a decreasing $E(m_\lambda)$ with wavelength with a value of 0.33 at 312.5 nm and 0.22 at 787.5 nm. This $E(m_\lambda)$ model was also assumed in the data analysis of [6].

In this section the ‘experimentally measured’ spectral radiation intensities were numerically simulated by Eq. (1), which includes the effect of self-absorption, using the known T and f_v distributions and the assumed $E(m_\lambda)$ from [6]. These numerically generated noise-free spectral radiation intensities were then used to reconstruct T and f_v distributions using the two methods described above and the direct Abel inversion of the spectral intensities without self-absorption correction. Effects of experimental error in the detected emission data on the inverted soot temperature and volume fraction distributions will be discussed later. The inverted T and f_v distributions at $z = 3, 4$ and 5 cm are compared with the input distributions in Figs. 2-4, respectively. These results were calculated using a spatial resolution of $\Delta r = \Delta y = 0.25$ mm based on the data given in [6]. Such a spatial resolution can be regarded as acceptable, though the sharp soot annular region shown in Fig. 2 is only marginally resolved. Although a higher spatial resolution is required to better resolve the soot volume fraction distribution in the annular region at $z = 3$ cm, such soot temperature and volume fraction profiles were not reported in [6]. The adequacy of this spatial resolution is also supported by the study of Ayrançi et al. [8] who investigated the effect of scanning resolution in a very similar laminar diffusion flame as [6] and found that the visible flame radius should be divided into no less than uniform 20 divisions to obtain accurate inverted soot temperature and volume fraction profiles. In our study the visible flame radius was divided into 17 uniform divisions using the soot temperature and volume fraction profiles of [6]. Nevertheless, the effect of scanning resolution (Δy in Fig. 1) or spatial resolution on the reconstructed soot temperature and volume fraction distributions was

investigated by reducing Δy by half and obtaining the input soot temperature and volume fraction values at the added y -locations through interpolation. For the noise-free input soot temperature and volume fraction profiles considered in this section, the effect of scanning resolution was found unimportant and the inverted results from the finer scanning resolution at the three heights are almost the same as those shown in Figs. 2 to 4.

In the following discussions the inverted soot temperature and volume fraction results obtained with and without self-absorption correction are to be compared with those of the input or assumed distributions. It is evident that the effect of self-absorption is somewhat significant and should be taken into account in this moderately sooting ethylene/air flame. At $z = 3$ cm where the soot volume fraction distribution exhibits a fairly strong annular structure, Fig. 2, neglect of self-absorption correction results in higher soot temperatures in the centerline region by about 10 to 15 K, but lower soot temperatures in the annular region of high soot volume fractions, by up to 31 K. Correspondingly, soot volume fraction is underestimated by about 10% in the centerline region but overestimated in the annular region of high soot volume fraction also by about 10%. At this flame height, the maximum optical thickness occurs at $y = 2.25$ mm and varies from 0.62 at 312.5 nm to 0.16 at 787.5 nm.

At $z = 4$ cm, Fig. 3, the soot volume fraction still displays an annular structure, peaks at a radial position of $r = 2$ mm, but is enhanced in the centerline region. The largest errors caused by neglect of self-absorption now occur in the centerline region with temperature being underestimated by 31 K while volume fraction overestimated by almost 16%. The errors in soot temperature (27 K) and volume fraction (9%) in the annular region of peak soot concentration around $r = 2$ mm are somewhat smaller than those in the centerline region, but still quite significant. As a result of higher soot concentrations in the flame centerline region at $z = 4$ cm,

the maximum optical thickness occurs along the line-of-sight passing through the flame center ($y = 0$) and varies from 0.61 at 312.5 nm to 0.16 at 787.5 nm. At this height, the inverted soot temperature is always lower than the input value when self-absorption correction is neglected.

At a higher location of $z = 5$ cm, Fig. 4, the annular structure of the soot volume fraction distribution nearly disappears and the peak concentration occurs in the centerline region at a much lower level of just above 4 ppm and the visible flame becomes much narrower as the visible flame tip is approached. At this height, the maximum optical thickness again occurs along the line-of-sight passing through the flame center ($y = 0$) and varies from 0.31 at 312.5 nm to 0.083 at 787.5 nm. These optical thicknesses are about half of those at $z = 4$ cm. As a result of the small optical thicknesses at $z = 5$ cm, the effects of self-absorption are fairly weak: the soot temperatures are again consistently underestimated by up to 14 K and soot volume fraction overestimated by about 6% in the centerline region.

The effects of self-absorption have been discussed previously by Snelling et al. [7]. Neglect of self-absorption influences the reconstructed T and f_v through two mechanisms. The first is the direct impact of self-absorption on the emission intensity governed by Eq. (1). It is evident that the degree of self-absorption is primarily dependent on the flame size (through the integration limits in the transmission term) and soot volume fraction and detection wavelength (through the soot absorption coefficient in the integration of the transmission term), and on the value of $E(m_\lambda)$ and how it varies with wavelength (as the uncertainty in $E(m)$ in the visible spectrum can be about 30%). Based on Eq. (2), the effect of self-absorption is expected to be stronger at a shorter detection wavelength. The wavelength dependence of the self-absorption effect, which was also recognized by Snelling et al. [7], suggests that self-absorption effect is determined by both the flame properties (size and soot loading levels) and how the emission

intensities are collected through the range of detection wavelength in the visible spectrum. In multi-wavelength flame emission measurement, the soot temperature is derived from the slope of the linear fit of $\ln[E(m_\lambda) \times S_\lambda(r)^{-1} \times \lambda^{-6}]$ vs. λ^{-1} . Self-absorption reduces the detected flame emission intensities, but non-uniformly across the flame spatially (different y values in Fig. 1) as well as non-uniformly spectrally. The local emission rates are also correspondingly underestimated if self-absorption were uncorrected, especially at lower wavelengths and the centerline and high soot concentration regions. At a given radial location the wavelength dependence of the self-absorption effect results in a larger slope of $\ln[E(m_\lambda) \times S_\lambda(r)^{-1} \times \lambda^{-6}]$ vs. λ^{-1} plot if the self-absorption effect were uncorrected, since the local emission rate is increasingly underestimated with decreasing wavelength, leading to a lower soot temperature. This is why the reconstructed soot temperatures are in general lower than the input values at the three heights shown in Figs. 2-4 when self-absorption is neglected. Therefore, the first influence of self-absorption is associated with its spectral dependence nature. However, Fig. 2 also shows that in the core of the flame the temperature is overestimated when self-absorption is neglected, which manifests the second influence of neglecting self-absorption. The second influence of self-absorption is associated with its spatial dependence, since at a given wavelength its effect is non-uniform along different line-of-sight characterized by its distance from the flame centre. When the attenuated emission intensities, which are detected experimentally, are inverted using an Abel inversion algorithm without taking into account self-absorption, erroneous local emission rate distribution can be produced due to the non-linear nature of the self-absorption effect, which is particularly true when the soot concentration displays a strong annular structure, i.e., the soot concentrations in the centerline region are very low such as at $z = 3$ cm. In this situation, inversion of the attenuated emission intensities leads to overestimation of the local emission rate in the centerline

region and the overestimation is more pronounced for shorter wavelengths. This is why the soot temperatures are somewhat overestimated in the centerline region at $z = 3$ cm. Such error is difficult to predict but evidently can lead to overestimation of temperature in the centre region, where the line-of-sight emission data experience the largest self-absorption effect, when the soot concentrations are very low, despite the general expectation of lower temperature discussed in the first mechanism.

The influence of attenuation on soot volume fraction is even more difficult to predict since the determined soot volume fraction is dependent on both the local emission rate and the local temperature, which are both influenced by self-absorption in a rather complex way, especially the former. However, some discussions are offered later with regard to the effect of different assumptions of the $E(m)$ vs. λ relationship on the inverted soot volume fraction.

Now examining the two self-absorption correction algorithms we see from Figs. 2-4 that at all three heights, the recovered T and f_v distributions using the Snelling et al. method [7] are almost identical to the input ones, suggesting it is very effective, regardless the optical thickness or the structure of the soot concentration (highly annular or not). At $z = 3$ cm, Fig. 2, the method of Freeman and Katz [23] is reasonably successful in correcting the self-absorption effect, since the reconstructed soot temperature and volume fraction distributions are in better agreement with the input distributions than those without self-absorption correction. This is particularly true around the annular region of high soot volume fractions. However, the improvements in the centerline region are insignificant. The inverted soot temperatures are overestimated in the centerline region by about 15 K, but underestimated by about 10 K in the annular region of high soot concentration. The inverted volume fractions remain about 10% lower in the centerline region and 4% higher in the peak soot concentration region. At $z = 4$ cm, Fig. 3, the Freeman and

Katz method reproduced more accurate T and f_v distributions than those at $z = 3$ cm, though the optical thicknesses at these two heights are very similar. It is also interesting to notice that the Freeman and Katz correction made significant improvements in both soot temperatures and volume fractions in the centerline region over those without self-absorption correction at $z = 4$ cm, but not at $z = 3$ cm. It is likely that the Freeman and Katz method is also less effective when the soot concentration displays a highly annular distribution and the optical thicknesses are large. At $z = 5$ cm, Fig. 4, where the maximum optical thickness is about 0.31, the Freeman and Katz correction method performs very well, as the reconstructed soot temperatures and volume fractions are in very good agreement with the input distributions across the flame at this height. This is indeed expected since this method is formulated using the linear approximation of the exponential function. At an optical thickness of 0.3, the linearization of the exponential function leads to an error of about 5.5% in transmissivity. Such error increases quickly to about 10.5% at a higher optical thickness of 0.4.

It is interesting to compare the results at $z = 3$ cm shown in Fig. 2 with regard to the effect of self-absorption correction with those obtained by Snelling et al. in a very similar flame (identical visible flame height, slightly different fuel tube diameters and fuel flow rates) at the same height shown in Fig. 6 of [7]. The results of Snelling et al. [7] at the flame height of $z = 3$ cm show that self-absorption correction only slightly increases the soot temperature by about 10 to 15 K almost uniformly except very close to the flame edge and has virtually no influence on the soot volume fraction. Their results on the effects of self-absorption are clearly different from the present results, though the very same self-absorption correction method is used. The potential causes for these different results lie in the different ranges of detection wavelength and different $E(m_n)$ models. In the study of De Iuliis et al. [6] and the present numerical work the detection

wavelength covers the range of 300 to 800 nm and a decreasing $E(m_\lambda)$ with wavelength is assumed. On the other hand, the emission intensities were detected between 500 and 945 nm (in two scan ranges: 500 to 748 nm and 685 to 945 nm) and a constant $E(m_\lambda)$ of 0.26 was used in the study of Snelling et al. [7]. Although both factors contribute to the different observations with regard to the self-absorption effect in [6] and [7], the main reason for their different findings lie in the assumption of the $E(m)$ vs. wavelength relationship in view of the results presented in this section and the next one.

3.2 *Effect of self-absorption for constant and increasing $E(m)$ with wavelength*

Results presented in Section 3.1 were obtained using the $E(m_\lambda)$ values of Chang and Charalampopoulos [28], which decrease with increasing wavelength in the visible spectrum. Other studies have arrived at different relationships between $E(m)$ and wavelength in the visible. Based on a comparison between multi-wavelength flame emission derived soot temperatures and those from CARS (coherent anti-Stokes Raman scattering) measurements Snelling et al. [7] suggested that a constant $E(m)$ over the spectral range of 500 to 945 nm is a good assumption. On the other hand, Krishnan et al. [29] found that $E(m)$ increases moderately with wavelength from about 0.22 at 350 nm to about 0.3 at 650 nm. Given such uncertainty in how $E(m)$ varies with wavelength in the visible, it is of importance to investigate how the $E(m)$ model affects the inverted soot temperature and volume fraction distributions if self-absorption effect were neglected. For this purpose, more calculations were conducted using constant $E(m)$ of different values and linearly increasing $E(m)$ with wavelength with $E(m) = 0.221$ at 312.5 nm and $E(m) = 0.316$ at 787.5 nm. The increasing $E(m)$ case was selected to be more or less opposite to the decreasing $E(m)$ one and also close to the values recommended by Krishnan et al. [29]. These

calculations were carried out for the same input soot temperature and volume fraction profile as those in the last section and for the spatial resolution of $\Delta y = 0.25$ mm.

Results obtained for constant $E(m)$ are shown in Fig. 5 for two $E(m)$ values of 0.2 and 0.32, which covers the range of decreasing $E(m)$ values with wavelength used in the last section, at the flame height of $z = 3$ cm. Results obtained at the other two flame heights, $z = 4$ and 5 cm, exhibit similar trends as those observed in Fig. 5, but at reduced differences between different treatments of self-absorption correction. Given the fairly significant effect of self-absorption shown in Figs. 2-4 under the assumption of decreasing $E(m)$ with wavelength, it is quite surprising to observe that the self-absorption correction has almost no influence on the inverted soot volume fraction in the annular high soot concentration region and the influence in the centerline region is very small for both values of $E(m)$. Although the self-absorption correction does affect the reconstructed soot temperature distribution, the influence is smaller than that in Fig. 2 even for the higher $E(m)$ value of 0.32. It is evident from Fig. 5 that the self-absorption effect under the assumption of constant $E(m)$ is greater for a larger value of $E(m)$, especially in the inverted soot temperature distribution, which is expected since a higher value of $E(m)$ corresponds to a higher value of absorption coefficient and therefore a larger optical thickness.

Figure 6 compares the reconstructed soot temperature and volume fraction distributions at $z = 3$ and 4 cm under the assumption of increasing $E(m)$ with wavelength. Although the effect of self-absorption on the inverted soot volume fraction distribution in this case is somewhat larger than that shown in Fig. 5 for constant $E(m)$, the effect is still significantly smaller than that for the increasing $E(m)$ case at both flame heights shown in Figs. 2 and 3. Unlike the case of assuming decreasing $E(m)$ with wavelength shown in Figs. 2-4 where neglect of self-absorption results in overestimation of soot volume fraction, assuming increasing $E(m)$ with wavelength

leads to underestimation of soot volume fraction. Although neglect of self-absorption in general leads to underestimation of soot temperature, as discussed earlier, the reconstructed soot volume fraction can be higher or lower than or even almost the same as the input values, depending on the assumption of how $E(m)$ varies with wavelength.

The results shown in Figs. 2-6 indicate that the self-absorption effect on the reconstructed soot temperature and volume fraction distributions is fairly sensitive to the assumption of how $E(m)$ varies with wavelength, which is somewhat unexpected. Here we present a brief discussion about the role of $E(m)$ vs. λ relationship to the self-absorption effect on the inverted soot volume fraction in an attempt to understand the results shown in Figs. 2-6. As given in Eq. 4, the local soot volume fraction is proportional to the inverted source term S_λ and inversely proportional to the local blackbody emission intensity $B_\lambda(T)$. When the effect of self-absorption is not corrected, both S_λ and $B_\lambda(T)$ are underestimated. The sign of the error in the soot volume fraction is then dependent on the relative deviations of S_λ and $B_\lambda(T)$ from the correct values. For example, if S_λ is more significantly underestimated than $B_\lambda(T)$, the resultant soot volume fraction will be lower than the correct value. Because the problem is highly nonlinear, it is difficult to predict how neglect of self-absorption correction would affect the inverted soot volume fraction, i.e., higher or lower than the correct value, for a given assumption of the $E(m)$ vs. λ relationship. However, it is useful to qualitatively understand how the $E(m)$ vs. λ relationship affects the inverted local source term S_λ and the local blackbody emission intensity $B_\lambda(T)$. For decreasing $E(m)$ with wavelength, the higher $E(m)$ value at a shorter wavelength makes an increasingly larger deviation of the local source term from the correct value with decreasing wavelength. Since the local temperature is determined by the slope of the $\ln[E(m_\lambda)/(\lambda^5 S_\lambda)]$ vs. λ^{-1} , the decreasing $E(m)$ with wavelength case is expected to cause the largest error in soot temperature if self-absorption

effect is not corrected, followed by the case of constant $E(m)$. The assumption of increasing $E(m)$ leads to the smallest error in soot temperature. The general effect of the $E(m)$ vs. λ relationship on the inverted source term at a given location is qualitatively illustrated in Fig. 7 based on the numerical results at $r = 2.75$ mm and $z = 3$ cm. It is evident from Fig. 7 that the slope of the uncorrected plot of the increasing $E(m)$ case deviates the least from the corrected one, leading to the least error in the inverted soot temperature, followed by the cases of assuming constant $E(m)$ and decreasing $E(m)$, as seen from results shown in Figs. 2-6. It seems that in the case of assuming constant $E(m)$ the percentage of error in the inverted local source term S_λ when self-absorption is uncorrected is compensated by a very similar percentage error in the local blackbody emission intensity, at least in the ethylene diffusion flame under consideration, leading to very small error in the soot volume fraction, Eq. 4. In the case of assuming decreasing $E(m)$ with wavelength, neglect of self-absorption effect results in too large error in soot temperature (lower than the correct value), due to the largest deviation in the slope of the curve shown in Fig. 7, compared to the error in the local source term. Therefore, the local soot volume fraction is in general overestimated under this assumption. On the other hand, assuming an increasing $E(m)$ with wavelength causes a very small deviation of the slope shown in Fig. 7 for the uncorrected plot, which means very small error in the derived local soot temperature. In this case, the local source term is underestimated to a greater degree than the underestimation of local soot temperature. This is why the soot volume fraction is underestimated compared to the correct value when an increasing $E(m)$ with wavelength is assumed.

Although it is useful to understand how the $E(m)$ vs. λ relationship can affect the reconstructed soot temperature and volume fraction distributions when self-absorption correction is not accounted for in the inversion of numerically simulated emission data using known soot

temperature and volume fraction profiles, it is important to point out that this situation does not correspond to the reality. In practical applications the soot temperature and volume fraction profiles are unknown and the available information is the line-of-sight spectral emission data. An $E(m)$ vs. λ relationship must be selected in the inversion of the emission data to recover the local soot temperature and volume fraction.

3.3 *Effect of the range of detection wavelength*

As noted earlier, the degree of self-absorption is strongly dependent on the detection wavelength and the value of $E(m_s)$ and how it varies with wavelength. From a comparison of the results of De Iuliis et al. [6] and Snelling et al. [7], where similar soot loadings and flame temperature are expected, it is evident that error due to self-absorption is more pronounced when using shorter detection wavelengths and decreasing $E(m)$ with wavelength, as in [6]. Although the spectral dependence of $E(m)$ is subject to uncertainty, the range of detection wavelength can be selected in flame emission measurements.

To explore the influence of spectral range on the self-absorption bias, numerical calculations were conducted using the input data at $z = 4$ cm. In these numerical calculations, the decreasing $E(m)$ with wavelength was used and the detection wavelength range was narrowed. It was found that narrowing the detection wavelength range to 600 to 800 nm, where the optical thickness varies between 0.22 at 600 nm and 0.16 at 800 nm, substantially reduces the error in the inverted soot temperature and volume fraction, while narrowing it to a lower range of 300 to 550 nm, where the optical thickness varies between 0.6 at 300 nm to 0.26 at 550 nm, significantly increases the error in the reconstructed soot temperature (38 K) and volume fraction (27%) if self-absorption were neglected. Therefore, it appears advantageous to detect flame

emission intensities at longer wavelengths to minimize the effect of self-absorption, in addition to offering better signal-to-noise ratios, further reducing the impact of scattering, and avoiding possible interferences from soot precursor materials, i.e., polycyclic aromatic hydrocarbons [30].

3.4 *Inversion of noisy emission intensities*

The results presented above were based on numerically calculated emission intensities with known soot temperature and volume fraction distributions, which correspond to an ideal situation where the emission intensities do not contain experimental errors. In reality, however, the detected emission intensities always contain errors or noise. To investigate how self-absorption correction affects the reconstructed soot temperature and volume fraction distributions when the emission intensities contain errors, numerical calculations were conducted by adding a certain amount of noise to the calculated emission intensities. In this study the calculated emission intensities were added by some noise whose magnitude is controlled by the product of the calculated emission intensity, the inverse of a specified signal-to-noise ratio (SNR), and a uniformly distributed random variable between -1 and 1. In these calculations, the $E(m)$ values based on the Chang and Charalampopoulos refractive index (decreasing with wavelength) were used and two SNR values of 500 and 100 were considered. In addition, calculations were conducted using two levels of spatial resolution, i.e., $\Delta y = 0.25$ mm and 0.125 mm. Unless otherwise stated, all the inverted soot temperature and volume fraction from noisy emission intensities in this work were obtained by adding specific noise distribution (using random numbers generated from specific seed values) to emission intensities.

The reconstructed soot temperature and volume fraction distributions at $z = 3$ cm for a moderate SNR of 500 and a low SNR of 100 are shown in Figs. 8 and 9 for the coarse and fine

spatial resolution, respectively. It is evident from these two figures that the reconstructed soot temperature and volume fraction display increasingly larger deviations from the input values with decreasing distance to the centerline ($r = 0$) and increasing level of noise in the emission intensities. This trend is well known for Abel inversion due to error accumulation. It is interesting to observe that the inverted soot temperatures and volume fractions from all three inversion methods in the outer region of the flame of $r \geq 1.5$ mm at $z = 3$ cm are essentially unaffected by the noise in the emission intensities, regardless of the level of noise or spatial resolution. The relative performance of the three inversions methods in the outer region of the flame remains almost the same as that observed in Fig. 2 for noise-free emission intensities. A comparison between Fig. 8 and Fig. 9 also indicates that it is advantageous to use the coarser spatial resolution in the inversion of noisy data to obtain smaller oscillations in the inverted soot temperature and volume fraction in the centerline region. Another important consideration in selecting the spatial resolution in the inverse analysis is that the spatial resolution should also be fine enough to adequately resolve the annular high soot concentration region. The results shown in Fig. 8(c) are the mean values of soot temperature and volume fraction averaged over the inverted results from the simulated noisy emission intensities using the inversion method of Snelling et al. and 30 sets of different seed values in the generation of random numbers (different seed values at different radial locations and different wavelengths) at SNR = 100. The error bars indicate the standard deviations. The standard deviations are quite small across the flame at this height, especially at radial locations greater than about $r = 1.5$ mm, except at the centerline where the deviations are very large. In addition, the mean values of soot temperature and volume fraction deviate from the input values in the centerline region, suggesting that direct inversion of noisy emission intensities can lead to systematic error in the inverted soot temperature and

volume fraction. Comparison between Figs. 8(b) and 8(c) suggests that the ‘instantaneous’ values of soot temperature and volume fraction inverted from the simulated noisy emission intensities by adding noise generated from a specific set of random numbers, Fig. 8(b), are very similar to the mean values, Fig. 8(c), except at the centerline. For this reason and the for sake of clarity, only the ‘instantaneous’ values of soot temperature and volume fraction inverted from simulated noisy emission intensities are considered in the rest of the paper.

Figure 10 displays the inverted soot temperature and volume fraction results for $\text{SNR} = 100$ and at $z = 5$ cm. Although a high level of noise is added to the emission intensities at this height, the inverted results with self-absorption correction accounted for, i.e., the Snelling et al. method or the Freeman and Katz method, are still consistently more accurate than those without self-absorption correction, indicating again that it is beneficial to account for self-absorption correction even in the inversion of noisy emission intensities. Again, use of the coarser spatial resolution gives rise to smaller errors in the centerline region. Overall, the errors in the inverted soot temperature and volume fraction at $z = 5$ cm due to the noise in the emission data are much smaller than those at $z = 3$ cm shown in Figs. 8 and 9. In practical applications of the multi-wavelength flame emission technique for soot temperature and volume fraction measurements, the detected line-of-sight emission data, which contain noise, should be preconditioned to filter out high-frequency noise by smoothing the experimental data before an inversion algorithm is applied [31]. Alternatively, the Abel inversion of noisy experimental data can be performed using the Tikhonov regularization method [32].

3.5 Analysis of the experimental data of De Iuliis et al.

Having assessed the effect of self-absorption, the role of $E(m)$ vs. λ relationship to the inverted soot temperature and volume fraction distribution when self-absorption is not corrected, and the impact of detection wavelengths, this section focuses on what the soot temperature and volume fraction distributions at flames heights of $z = 3$ and 4 cm would be if self-absorption correction were accounted for in the data inversion. Since the measured multi-wavelength flame emission intensities of De Iuliis et al. [6] are unavailable and their data inversion was performed with self-absorption neglected, it is reasonable to assume that their measured spectral intensities correspond to those calculated by the reported T and f_v distributions given in Figs. 5 and 3 of [6] and Eq. (5), i.e., without self-absorption. We will use such calculated spectral intensities to reconstruct T and f_v distributions at $z = 3$ and 4 cm by the method of Snelling et al. to demonstrate what the reconstructed T and f_v would be if self-absorption were accounted for in their data analysis. The results are shown in Figs. 11 and 12 using the decreasing $E(m)$ with wavelength of Chang and Charalampopoulos [28]. As expected from the results shown earlier in Figs. 2 and 3, inclusion of self-absorption correction in the data inversion leads to noticeably higher soot temperatures in the centerline region at $z = 4$ cm and the high soot concentration region at $z = 3$ cm by up to 30 K. The reconstructed soot volume fractions with self-absorption correction are in general lower than the values reported in [6] by up to 12%.

3.6 Analysis of the flame emission data of Thomson et al.

The spectrally resolved flame emission data measured by Thomson et al. [13] in a laminar coflow methane/air diffusion flame at 40 atm at four flame heights of $z = 2, 3, 4,$ and 5 mm were reanalyzed. This flame is very small (only about 2 mm in diameter and about 8 mm tall) but has a very high level of soot loading at more than 100 ppm. The spectral flame emission

intensities were detected over a spectral range of 700 to 933 nm and binned at a uniform spectral resolution of 21 nm. Following [13], a constant value of $E(m_\lambda)$ of 0.274 was used in the analysis.

The differences between the reconstructed T and f_v distributions with and without self-absorption correction at the four heights are qualitatively similar. Only the results at $z = 2$ and 4 mm are shown in Figs. 13 and 14, respectively. The optical thickness across the flame center varies from 0.44 (700 nm) to 0.33 (930 nm) at $z = 2$ mm and from 0.55 (700 nm) to 0.41 (930 nm) at $z = 4$ mm. These optical thicknesses are somewhat smaller than, but similar to, those at the shorter detection wavelengths and at $z = 3$ and 4 cm of the ethylene/air diffusion flame studied by De Iuliis et al. [6]. When self-absorption is neglected, the soot temperature is in general underestimated by up to 50 K at $z = 2$ mm and up to 64 K at $z = 4$ mm. The effect of self-absorption on the reconstructed soot temperature in this high-pressure methane/air flame is hence somewhat stronger than that in the ethylene/air diffusion flame shown in Figs. 11 and 12. On the other hand, the effect of self-absorption on the reconstructed soot volume fraction distribution is fairly small at both flame heights, being less than 10% in the centerline region at $z = 4$ mm, Fig. 13, and in general less than 5%. The present reconstructed soot volume fraction distributions without self-absorption correction are in good agreement with those obtained by Thomson et al. [13], as expected, since the present analysis was conducted using the same inversion method to the same experimental data as in [13]. Overall, it can be seen that the soot temperatures reported in [13], where the effect of self-absorption was neglected, are underestimated by up to about 60 K, especially in the annular regions where the soot loading is high and soot concentration falls sharply due to oxidation. The inverted soot temperatures with self-absorption corrected accounted for using the Freeman and Katz method are very close to, but slightly lower than, those using the Snelling et al. method in the outer region at both flame heights. In the centerline

region, however, the opposite trend is observed, especially at $z = 4$ mm, Fig. 14. The inverted soot volume fractions using the Freeman and Katz method are in close agreement with those from Snelling et al. method.

The flame emission data of Thomson et al. [13] were also analyzed using the spectral dependent $E(m_\lambda)$ in [28]. It was found that while the reconstructed T and f_v differ from those obtained using a constant $E(m_\lambda)$ of 0.274, especially soot volume fraction, the effect of self-absorption remains almost unchanged. This is because over the relatively narrow spectral range of 700 to 930 nm the value of $E(m_\lambda)$ decreases only slightly from 0.223 at 700 nm to 0.218 at 930 nm.

3.7 *Effect of optical thickness*

Although the results presented above clearly indicate that self-absorption does affect the reconstructed soot temperature and volume fraction distributions in laminar diffusion flames, its overall influence is considered relatively small to moderate compared to the uncertainties in the soot absorption function $E(m)$, which can be as large as about 30%. Since self-absorption effect becomes more significant in flames of larger optical thicknesses, it is more important to account for this effect in the flame emission data inversion from large sooting flames, such as pool fires. To demonstrate the importance of self-absorption effect in flames of large optical thickness and to illustrate the effectiveness of the Snelling et al. method in such flames, more calculations were conducted for higher soot loadings by multiplying the input soot volume fraction profiles in Section 3.1 by a factor of 4 and 10 while keeping the soot temperature profiles unchanged. It was found from numerical calculations that the inverted soot volume fraction becomes very sensitive to the accuracy of the inverted soot temperature in the Snelling et al. method at the highest soot

loading considered. To obtain smooth soot volume fraction distribution using the Snelling et al. method the tolerance of soot temperature iteration was reduced to 2×10^{-5} from 2×10^{-4} when soot volume fraction in Section 3.1 was increased by a factor of 10.

Figure 15 shows the results obtained for raising the input soot volume fractions in [6] by a factor of 4 using the different assumptions of the $E(m)$ vs. λ relationship at the flame height of $z = 3$ cm. The results shown in Fig. 15 were calculated without adding noise to the simulated emission data over the spectral range of 300 to 800 nm. The optical thicknesses along the path through the soot annular region vary between about 2.5 to 0.6 at 312.5 nm and 787.5 nm, respectively, depending on the $E(m)$ assumptions. It is evident that the self-absorption effect is very significant at this enhanced soot loading with errors in soot temperature and volume fraction being as large as 110 K and 60%, respectively. The largest errors caused by neglect of self-absorption occur under the assumption of decreasing $E(m)$. The Snelling et al. method is very effective in recovering accurately both soot temperature and volume fraction. As expected, the Freeman and Katz method loses its effectiveness at such high optical thicknesses. As discussed earlier in Section 3.1, soot temperatures are seen in general to be underestimated if self-absorption were neglected, though overestimation can occur in the centerline region for highly annular distributions of soot concentration, such as at $z = 3$ cm. The inverted soot volume fractions can be either higher or lower than the input values, depending on the assumption of the $E(m)$ vs. λ relationship. The effects of different $E(m)$ assumptions on the reconstructed soot temperature and volume fraction distributions when self-absorption correction is not accounted for have already been discussed in Section 3.2. The general trends observed from results shown in Figs. 2-6 with regard to the role played by the $E(m)$ assumption can also be seen from results in Fig. 15 for the higher soot loading case, though the deviations between the uncorrected and

corrected results for self-absorption become much larger. The main difference between the results shown in Fig. 15(b) and Fig. 5 under the assumption of constant $E(m)$ lie in the inverted soot volume fractions. At the low soot loading, Fig. 5, self-absorption correction has negligible influence on the inverted soot volume fractions in the annular high soot concentration region. This is no longer true in the higher soot loading case, Fig. 15(b), where neglect of self-absorption correction results in about 7% lower in the inverted soot volume fractions in the annular region.

Results for the highest soot loading considered using the assumption of increasing $E(m)$ with wavelength are shown in Figs. 16-18 for $z = 3$, 4, and 5 cm, respectively. In these calculations the simulated emission data over the spectral range of 300 to 800 nm were added by noise with SNR = 400. The maximum optical thickness in this case reaches 4.92 at 312.5 nm in the annular region at $z = 3$ cm. Even in this optically thick case with noise in the emission data, the Snelling et al. method is still capable of recovering accurately the soot temperature and volume fraction distributions at all three flame heights, except in the centerline region low in the flame at $z = 3$ and 4 cm. Neglect of self-absorption leads to very large errors in both soot temperature and volume fraction, with the latter being greater than 100%, Figs. 16 and 17. Although the Freeman and Katz method recovers better soot volume fractions and temperatures, except in the centerline regions at $z = 3$ and 4 cm, Figs. 16 and 17, than those without self-absorption correction, it in general still produces very large errors.

Results shown in Figs. 15-18 demonstrate that self-absorption effect can be very important in large scale sooting flames and it is crucial to correct this effect in the inversion of spectral flame emission measurements. These results also indicate that the self-absorption correction method of Snelling et al. [7] is accurate even in the inversion of spectral flame emission data for optical thicknesses as large as 5. However, it should be recognized that when

the flame optical thickness is sufficiently large the emissions originated from the centerline region are so severely attenuated by the outer soot containing region that they essentially cannot reach the detector. In this situation, the flame emission technique cannot resolve soot temperature and volume fraction distributions based on detection of flame emissions.

4. Conclusions

The effect of self-absorption on the retrieval of soot temperature and volume fraction distributions in axisymmetric laminar coflow diffusion flames in multi-wavelength flame emission measurements was investigated using the methods of Freeman and Katz [23] and Snelling et al. [7]. The effect of self-absorption should be accounted for when the maximum optical thickness of the flame exceeds 0.3. The method of Freeman and Katz is reasonably accurate as long as the flame thickness remains less than about 0.4 and the soot distribution is not highly annular. The method of Snelling et al. is very accurate in recovering the unattenuated line-of-sight spectral emission intensities based on the measured attenuated data, regardless of the flame optical thickness (as large as 5), the structure of soot concentration distribution, noise in the emission data, or the relationship between the soot absorption function and wavelength. Spatial resolution affects the errors in the inverted soot temperature and volume fraction distributions in the centerline region. Use of a finer resolution produces larger errors in the reconstructed soot properties from the inversion of noisy emission data. An adequate spatial resolution is preferred to resolve the sharp gradients in the annular high soot concentration region while not to excessively magnify the error in the emission data in the centerline region. For spectral emission data containing large random errors, preconditioning of the noisy data is necessary to filter out the high-frequency noise before applying an inversion algorithm. As far as

numerical calculations are concerned, the method of Freeman and Katz provides no advantages over the method of Snelling et al. and is in general much less accurate.

Accounting for the effect of self-absorption in the analysis of the flame emission data of De Juiis et al. [6] obtained in a laminar coflow ethylene/air diffusion flame at atmospheric pressure indicates that their published soot temperatures were underestimated by about 30 K, while soot volume fractions were overestimated by about 12%. Analysis of the flame emission data of Thomson et al. [13] measured in a laminar coflow methane/air diffusion flame at 40 atm with and without the self-absorption correction shows that the flame temperatures were underestimated by about 60 K if self-absorption effect were neglected. However, the effect of self-absorption has a minor influence on the reconstructed soot volume fraction.

Neglect of self-absorption in general leads to lower soot temperatures, regardless of the detection wavelength range or the wavelength dependence of the soot absorption function assumed in the analysis, but it can also result in overestimate of temperature in the centerline region when the soot concentration distribution is highly annular. The assumed $E(m)$ vs. wavelength relationship affects the reconstructed soot temperature and volume fraction distributions when self-absorption is neglected. Compared to the assumptions of constant $E(m)$ or increasing $E(m)$ with wavelength, the assumption of decreasing $E(m)$ with wavelength results in the largest errors in the inverted soot temperature and volume fraction when self-absorption is not accounted for. Assumption of constant $E(m)$ leads to the least error in soot volume fraction, while the assumption of increasing $E(m)$ with wavelength gives rise to the smallest error in the inverted soot temperature.

The self-absorption effect is also dependent on the spectral range of the flame emission detection. It is advantageous to detect flame emission intensities in longer wavelengths in the

visible spectrum to minimize the effect of self-absorption if its effect were not accounted for in the data inversion. Since the Snelling et al. method is found effective in accounting for the self-absorption effect in the retrieval of soot temperature and volume fraction distributions even when noise is present in the emission data and it is fairly easy to implement, it is recommended to be used in the retrieval of soot temperature and volume fraction distributions in multi-wavelength flame emission measurements of axisymmetric sooting flames.

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